

ON UNIVERSALITY OF CONDUCTIVITY OF PLANAR RANDOM SELF-DUAL SYSTEMS

S.A.Bulgadaev ¹

Landau Institute for Theoretical Physics
Chernogolovka, Moscow Region, Russia, 142432

General properties of the effective conductivity σ_e of planar isotropic randomly inhomogeneous two-phase self-dual systems are investigated. A new approach for finding out σ_e of random systems based on a duality, a series expansion in the inhomogeneous parameter z and additional assumptions, is proposed. Two new approximate expressions for σ_e at arbitrary values of phase concentrations are found. They satisfy all necessary inequalities, symmetries, including a dual one, and reproduce known results in various limiting cases. Two corresponding models with different inhomogeneity structures, whose σ_e coincide with these expressions, are constructed. First model describes systems with a finite maximal characteristic scale of the inhomogeneities. In this model σ_e is a solution of the approximate functional equation, generalizing the duality relation. The second model is constructed from squares with random layered structure. The difference of σ_e for these models means a nonuniversality of the effective conductivity even for binary random self-dual systems. The first explicit expression for σ_e can be used also for approximate description of various inhomogeneous systems with compact inclusions of the second phase. The percolation problem of these models is briefly discussed.

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1. Introduction

The electrical transport properties of the classical inhomogeneous condensed media, being a particular case of the transport properties in disordered systems, have a great practical interest. For this reason they have been intensively studied theoretically as well as experimentally (see, for example, [1, 2]). An importance of this knowledge has revived last time. It was established that many real systems, which have been considered to be homogeneous, can have on small scales (from microscopic through mesoscopic till macroscopic) inhomogeneous or even heterophase structures. In particular, such situation takes place in the oxide materials, which are now actively studied [3]. Analogous situation takes place in many real examples of the disordered systems (for example [4]). For this reason the observed effective conductivity σ_e of these systems can differ from that of homogeneous media. Thus one needs a theory adequately describing conducting properties of such inhomogeneous media. Unfortunately, the theory of strongly inhomogeneous systems is not well developed due to a difficulty of explicit averaging over strong inhomogeneous fluctuations. The various perturbation, approximate and variation theories have been developed

¹e-mail: bulgad@itp.ac.ru

and many interesting results have been obtained so far (see, for example, [5] and a recent review [6]).

In the conducting properties of the inhomogeneous media there is one old problem about the effective conductivity σ_e of inhomogeneous (regular, irregular or random) heterophase systems, containing $N(N \geq 2)$ various phases with different partial conductivities σ_i ($i = 1, 2, \dots, N$). In general, the inhomogeneities can be very different, changing from regular through weakly and moderately irregular till strongly irregular ones. Strong inhomogeneous fluctuations can be connected with large differences of the partial conductivities as well as with strong irregularities of these phases. The latter situation is very similar to the case of random inhomogeneous medium and in some cases can be identified with it.

In principle, the regular and weakly irregular cases can be considered systematically in framework of classical potential theory [7], while an investigation of strong and random inhomogeneous systems needs some approaches connected with the averaging over the random regions with different conductivities [8].

The basic equation, describing a stationary electric current in random systems, is the Ohm law, connecting the local current $\mathbf{j}(\mathbf{r})$ and the local electric field $\mathbf{E}(\mathbf{r})$,

$$\mathbf{j}(\mathbf{r}) = \sigma(\mathbf{r}) \mathbf{E}(\mathbf{r}), \quad (1)$$

where $\sigma(\mathbf{r})$ is a local conductivity. We will assume that the carriers in all phases have the charges of the same sign. Then one can suppose that $\sigma(\mathbf{r}) \geq 0$. The equation (1) must be supplemented with the corresponding boundary conditions on the boundaries of two phases [1]

$$j_n^1 = j_n^2, \quad E_t^1 = E_t^2, \quad (2)$$

where n, t denote the normal and tangent components and 1, 2 correspond to different phases. The vector fields $\mathbf{E}(\mathbf{r})$ and $\mathbf{j}(\mathbf{r})$ satisfy the following equations inside each phase

$$\nabla \times \mathbf{E}(\mathbf{r}) = 0, \quad \nabla \cdot \mathbf{j}(\mathbf{r}) = 0. \quad (3)$$

They mean that $\mathbf{E}(\mathbf{r})$ and $\mathbf{j}(\mathbf{r})$ are potential and divergenceless fields respectively.

The effective conductivity σ_e of the isotropic randomly and/or strongly inhomogeneous medium can be defined as a proportionality coefficient between mean values of \mathbf{j} and \mathbf{E} averaged over the area of the system

$$\bar{\mathbf{j}} = \sigma_e \bar{\mathbf{E}}, \quad (4)$$

$$\bar{\mathbf{j}} \equiv \int \mathbf{j}(\mathbf{r}) d\mathbf{r} / S, \quad \bar{\mathbf{E}} \equiv \int \mathbf{E}(\mathbf{r}) d\mathbf{r} / S \quad (5)$$

where S is an area of the system.

Since it is very difficult to make an averaging of (4,5) over an area of the system explicitly for strongly irregular or random systems it is usually supposed that for systems with such $\sigma(\mathbf{r})$ (or with a fixed random realization of $\sigma(\mathbf{r})$) the

averaging of (4,5) over the area of the system can be replaced by the averaging over some ensemble of random functions $\sigma(\mathbf{r})$ with a functional weight $\mathcal{P}\{\sigma\}$, which may, in general, depend also on temperature T

$$\int d\mathbf{r} F(\sigma(\mathbf{r}))/S = \int \mathcal{D}\sigma(\mathbf{r}) F(\sigma(\mathbf{r})) \mathcal{P}\{\sigma\} \equiv \ll F(\sigma(\mathbf{r})) \gg . \quad (6)$$

In this paper we confine ourselves with an investigation of the σ_e dependence on σ_i and assume that T is fixed.

According to the general principles, a behaviour of the effective conductivity of random systems must depend on statistical properties of the random conductivity $\sigma(\mathbf{r})$ encoded in its correlations. In case of random heterophase systems one usually supposes that $\sigma(\mathbf{r})$ at different space points are statistically independent and its moments in each space point \mathbf{r} are defined by one distribution function of partial conductivities $P(\sigma)$, which does not depend on \mathbf{r} ,

$$P(\sigma) = \sum_{i=1}^N x_i \delta(\sigma - \sigma_i), \quad \sum_1^N x_i = 1, \quad \int P(\sigma) d\sigma = 1, \quad (7)$$

where x_i is the concentration of the i-th phase.

This means that a mean conductivity $\ll \sigma \gg$ (and all its higher moments) can be represented in two equivalent forms

$$\ll \sigma \gg = \int \sigma(\mathbf{r}) d\mathbf{r}/S = \int \sigma P(\sigma) d\sigma = \langle \sigma \rangle. \quad (8)$$

where $\langle \dots \rangle$ denotes the average with a distribution function $P(\sigma)$. Then the bare phenomenological correlation function of conductivities $C_0(r)$ takes the simple form

$$C_0(r, r') = \ll \sigma(r)\sigma(r') \gg = \langle \sigma \rangle^2 + \langle \sigma^2 \rangle \delta(r - r') \quad (9)$$

As follows from (9) $C_0(r, r')$ in this form does not already describe a detailed spatial structure of random inhomogeneities, but contains only their integral characteristics – the concentrations of phases. In this case σ_e can be represented as a function of partial conductivities σ_i and concentrations x_i only

$$\sigma_e = \sigma_e(\sigma_1, x_1 | \sigma_2, x_2 | \dots | \sigma_N, x_N) \quad (10)$$

Due to the linearity of the defining equations this function must be a homogeneous function of degree one of $\sigma_i, i = 1, \dots, N$. It has also satisfy the inequalities following from general principles (see, for example, [6])

$$\langle \sigma^{-1} \rangle^{-1} \leq \sigma_e \leq \langle \sigma \rangle. \quad (11)$$

For practical purposes and for a description of experimental data one needs usually some explicit and compact (though, maybe, approximate) formulas for effective conductivity applicable in a wide range of parameters. A general expression

for σ_e of isotropic media at arbitrary concentrations x_i can be obtained only in case of weakly inhomogeneous media, when the conductivity fluctuations $\Delta\sigma$ ($\Delta\sigma = \sigma - \langle\sigma\rangle$, are much smaller than a mean conductivity $\langle\sigma\rangle$ [1]

$$\sigma_e = \langle\sigma\rangle \left(1 - \frac{\langle(\delta\sigma)^2\rangle}{D}\right), \quad \delta\sigma = \frac{\Delta\sigma}{\langle\sigma\rangle}, \quad \langle(\Delta\sigma)^2\rangle = \langle\sigma^2\rangle - \langle\sigma\rangle^2, \quad (12)$$

where D is a dimension of the system. This formula expresses σ_e only through two first moments of random variable σ . The further theoretical results in this problem for random inhomogeneous systems are related with the effective medium approximation (EMA) [9] and the random resistor networks (RRN) models of these systems [2], since they allow to fulfil an averaging procedure explicitly. The RRN models approximate the continuous inhomogeneous systems (IS) and can be obtained as a result of some their discretization (though, in general, this procedure is not exact and unambiguous). It was shown for the RRN models firstly by numerical simulations [2] and later by series expansion on moments of $\delta\sigma$ [10, 11] that the EMA, dealing ad hoc with weakly IS, works well even for moderately inhomogeneous RRN models.

It is widely believed that the EMA, due to its good description of RRN type models, is also a good approximation for continuous random IS. As is known the EMA for different isotropic RRN models (for example, on isotropic regular lattices) gives different expressions for σ_e [2]. Thus σ_e in the EMA is not an universal function even for isotropic RRN models. At the same time, not every continuous random IS can be described by the discrete RRN models, since their reduction to the RRN type models can change some properties. Moreover, the continuous random IS can have more complicate local structures (see below). Then σ_e of such continuous systems can differ in higher order terms in $\delta\sigma$ (starting with the 3-rd order, because the two first terms are universal and are determined by the formula (12)) from the EMA. For example, it is not obvious that any system, containing random "resistors" and "conductors" (i.e. the elements inverted to each other) simultaneously, must have the same behaviour as a "pure" RRN model. For a description of continuous IS it is very desirable to have any method, basing on the first principles and general properties, which is not connected with the RRN type models and/or with the EM approximation. Fortunately, for two-dimensional systems, due to the existence of the exact duality relation [12, 13], one can attempt, basing on this relation, to find other possible approximations different from the EMA and applicable for systems with other inhomogeneity structures. In this paper we will show for the binary systems ($N = 2$) how starting with a duality relation (DR) and using some additional assumptions one can find two explicit approximate expressions for σ_e , differing from the EMA. We will also construct two corresponding models, having their effective conductivities just of these two forms, and discuss their properties and possible applications. A case of arbitrary number of phases N will be presented in the subsequent paper.

2. Reciprocity and duality relations and symmetries

We begin with a consideration of main general properties of σ_e , because in the existing literature there are some misunderstandings about them. For systems with δ -correlated inhomogeneities σ_e can be reduced to a function, depending on the pairs of partial parameters: conductivities σ_i and phase concentrations x_i ($0 \leq x_i \leq 1$, $i = 1, 2$, $\sum_1^2 x_i = 1$)

$$\sigma_e = \sigma_e(\sigma_1, x_1 | \sigma_2, x_2). \quad (13)$$

Note, that in general case the order of the arguments pairs is important, since the phases can have different geometrical and/or statistical properties. In two dimensions there is a reciprocity between divergenceless and curl free vector fields \mathbf{v} and $\tilde{\mathbf{v}}$

$$v_i = \epsilon_{ik} \tilde{v}_k, \quad \partial_i v_i = \epsilon_{ik} \partial_k \tilde{v}_i = 0. \quad (14)$$

This relation is local and takes place in each point. It means that the reciprocal vector field is the initial field turned on $\pi/2$. For this reason for continuous inhomogeneous (and isotropic) media the reciprocal vector field feels the same inhomogeneities of the initial system. Inside any phase one can connect with the reciprocal vector fields $\tilde{\mathbf{j}}$ and $\tilde{\mathbf{E}}$ a reciprocal conducting system, such that $\mathbf{j}_r \sim \tilde{\mathbf{E}}$ and $\mathbf{E}_r \sim \tilde{\mathbf{j}}$, and for which the corresponding Ohm law takes place

$$\mathbf{j}_r = \tilde{\sigma} \mathbf{E}_r, \quad \tilde{\sigma} = \sigma_0^2 / \sigma, \quad (15)$$

where σ_0 is some arbitrary constant with dimension of conductivity. It means that $\tilde{\sigma}$ is inversely proportional to σ and the coordinate axis of the reciprocal system are turned on $\pi/2$. Note that the corresponding phase boundary conditions (2) are also fulfilled, since the normal vectors transform into tangent ones and vice versa. The effective conductivities of the initial medium and its reciprocal must satisfy after averaging the same functional relation as in (15), which can be called the reciprocity relation (RR) and is usually written in the following form

$$\sigma_e(\sigma_1, x_1 | \sigma_2, x_2) \tilde{\sigma}_e(\sigma_0^2 / \sigma_1, x_1 | \sigma_0^2 / \sigma_2, x_2) = \sigma_0^2. \quad (16)$$

In this general form it has been obtained firstly by Keller [12] and independently by Dykhne [13]. The general RR (16) takes place for all two-dimensional binary systems: for RRN models and for regular and irregular media, as well as for random ones. But the general RR is not so useful, since it connects the effective conductivities of different systems. In order to obtain more information about σ_e one needs to know how these effective conductivities are related. The most interesting case is when a reciprocal system has the same statistical or/and geometrical properties. In this case the effective conductivity of a reciprocal system must coincide with the effective conductivity of the initial one. Such systems can be called the self-reciprocal ones (we will use a name self-dual for more symmetrical systems, see below). One can see from the above description of the reciprocal medium that for isotropic continuous random inhomogeneous medium the reciprocal medium will have the same type of inhomogeneities and after averaging its effective conductivity $\tilde{\sigma}_e$ must have the same functional form as the effective conductivity of the initial medium, but

with inversed partial conductivities. Thus they have satisfy the self-reciprocity relation (SRR)

$$\sigma_e(\sigma_1, x_1 | \sigma_2, x_2) \sigma_e(\sigma_0^2 / \sigma_1, x_1 | \sigma_0^2 / \sigma_2, x_2) = \sigma_0^2. \quad (17)$$

At the same time it is worth to note here that in case of discrete RRN type models the reciprocal network can differ from the initial one and consequently a functional form of the effective conductivity of the reciprocal network will differ from that of initial network. In particular, the effective conductivities of RRN models on isotropic regular lattices (let us call them random resistor lattice (RRL) models), triangle and honeycomb, turn out different and must satisfy, due to a mutual reciprocity of these lattices, only the R-relation

$$\sigma_e^{tr}(\sigma_1, x_1 | \sigma_2, x_2) \sigma_e^{hon}(\sigma_0^2 / \sigma_1, x_1 | \sigma_0^2 / \sigma_2, x_2) = \sigma_0^2, \quad (18)$$

This is a payment for an ambiguity of the discretization procedure of the continuous medium.

The self-reciprocal systems include the continuous media and RRN model on self-reciprocal networks. For self-reciprocal systems the SRR can be rewritten also in the form

$$\sigma_e(\sigma_1, x_1 | \sigma_2, x_2) = \sigma_0^2 / \sigma_e(\sigma_0^2 / \sigma_1, x_1 | \sigma_0^2 / \sigma_2, x_2). \quad (19)$$

This form has a simple physical sense, connected with two possibilities of a writing the Ohm law in terms of conductivities σ (as (15)) and in terms of resistivities $\rho = 1/\sigma$: an effective conductivity of a self-reciprocal system as a function of partial conductivities must coincide with an inversed resistivity as a function of corresponding partial resistivities (i.e. inversed partial conductivities). For this reason the relation (19) can be also called the inversion relation (IR).

Though now the SRR (17) restricts a possible functional form of σ_e , in a general case it does not yet permit to find the exact values for σ_e even at equal phase concentrations $x_1 = x_2 = 1/2$ and $\sigma_0^2 = \sigma_1 \sigma_2$ (the latter choice turns the inversion $\sigma \rightarrow \sigma_0^2 / \sigma$ into permutation $\sigma_1 \leftrightarrow \sigma_2$). To be able to find exact value of σ_e one needs additional symmetry property of systems connected with the geometrical and/or statistical properties of their partial phases. To see various possibilities, appearing for inhomogeneous media, let us consider a simple, but very instructive, case of regular inhomogeneous system consisting from one (white) phase with regular round inclusions of the second (black) phase, the Rayleigh model. This system is self-reciprocal in the above sense and its effective conductivity satisfies the SRR (and IR) (17) ([14]). However, the geometrical properties of these phases are different. The four pictures of this model at the interchanged phases and at the adjoint concentrations x and $1 - x$ of black phase are represented on Fig.1. As follows from Fig.1, all these examples of the model are different and, consequently, the corresponding σ_e are also different. Thus, though the SRR takes place for this model, it does not give any equation for σ_e . Such equation can be obtained only when σ_e is a

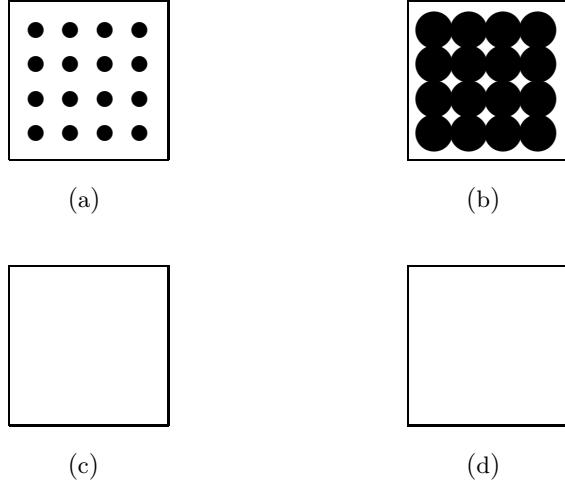


Figure 1: (a) A part of the model with regularly situated round inclusions at the concentration x , (b) a part of the same model at adjoint concentration $1 - x$, (c) the same model at concentration x but with the interchanged phases, (d) the same model with interchanged phases at adjoint concentration $1 - x$.

symmetrical under simultaneous interchange of both parameters of phases

$$\sigma_e(\sigma_1, x_1 | \sigma_2, x_2) = \sigma_e(\sigma_2, x_2 | \sigma_1, x_1). \quad (20)$$

This property can take place as for random systems as for some regular systems. The typical examples of such systems are the random mixtures and RRN models with the probability distribution function $P(\sigma)$ (7), and a regular "chess-board" type system. One can see from Fig.1 that the self-reciprocal Rayleigh model does not have this symmetry, but this symmetry appears as an approximate symmetry, when a concentration of one phase is small (then fig.1a (fig.1b) is similar to fig.1d (fig.1c)).

We will call symmetrical self-reciprocal systems as self-dual (SD) systems. The corresponding SRR takes at $\sigma_0^2 = \sigma_1\sigma_2$ the following form

$$\sigma_e(\sigma_1, x_1 | \sigma_2, x_2)\sigma_e(\sigma_1, x_2 | \sigma_2, x_1) = \sigma_1\sigma_2. \quad (21)$$

For SD systems one can obtain exact equation for σ_e . It takes place only at equal phase concentrations $x_i = 1/2$ and at $\sigma_0^2 = \sigma_1\sigma_2$. and gives the well-known exact Keller-Dykhne value [12, 13]

$$\sigma_e(\sigma_1, 1/2 | \sigma_2, 1/2) = \sqrt{\sigma_1\sigma_2}. \quad (22)$$

This remarkable formula is very simple and universal in a sense that it takes place for all two-dimensional inhomogeneous self-dual systems as for regular as for random.

For binary systems one can write σ_e in a more short form as a function of its independent parameters only (here x is a concentration of the first phase)

$$\sigma_e = \sigma_e(x, \sigma_1, \sigma_2). \quad (23)$$

Then, for isotropic SD systems with unequal phase concentrations (21) can be written in the form, which connects the effective conductivities at adjoint concentrations x and $1-x$ or in terms of a new variable $\epsilon = x - 1/2$ ($-1/2 \leq \epsilon \leq 1/2$) at ϵ and $-\epsilon$

$$\sigma_e(x, \sigma_1, \sigma_2) \sigma_e(1-x, \sigma_1, \sigma_2) = \sigma_1 \sigma_2 \quad (24)$$

Eq.(24) means that a product of the effective conductivities at adjoint concentrations is an invariant. Due to this relation one can consider σ_e only in the regions $x \geq 1/2$ ($\epsilon \geq 0$) or $x \leq 1/2$ ($\epsilon \leq 0$). Just the relations (21) and (24) are usually named as the duality relation (DR). Below we will consider mainly systems, satisfying the DR (21) or (24) as the exact functional relation. It follows also from (24) that (for $\sigma_1 \geq \sigma_2$) than faster is σ_e increasing at $x \geq 1/2$, than slower it increase at $x \leq 1/2$.

The binary case assumes further simplifications. It is convenient to use instead of $\sigma_i, i = 1, 2$ another combinations of σ_i : $\sigma_{\pm} = (\sigma_1 \pm \sigma_2)/2$. Then, the effective conductivity can be represented in the form, symmetrical relatively to both phases,

$$\sigma_e(\epsilon, \sigma_+, \sigma_-) = \sigma_+ f(\epsilon, \sigma_-/\sigma_+) = \sigma_+ f(\epsilon, z), \quad (25)$$

where a new dimensionless variable $z = \sigma_-/\sigma_+$, ($-1 \leq z \leq 1$), characterizing an inhomogeneity of the system, is introduced. This choice of variables is convenient for a study of σ_e in the limit, when one partial conductivity becomes small or goes to 0.

The effective conductivity $\sigma_e(\epsilon, \sigma_+, \sigma_-)$ and a function $f(\epsilon, z)$ must have the next boundary values

$$\begin{aligned} \sigma_e(1/2, \sigma_+, \sigma_-) &= \sigma_1, & \sigma_e(-1/2, \sigma_+, \sigma_-) &= \sigma_2, \\ f(1/2, z) &= 1 + z, & f(-1/2, z) &= 1 - z, & f(\epsilon, 0) &= 1. \end{aligned} \quad (25')$$

The duality relation takes in these variables the form

$$f(\epsilon, z) f(-\epsilon, z) = 1 - z^2, \quad (26)$$

from which one obtains at critical concentration $\epsilon = 0$ the exact value

$$f(0, z) = \sqrt{1 - z^2}. \quad (26')$$

The self-reciprocity relation in these terms has the form

$$f(\epsilon, z) f(\epsilon, -z) = 1 - z^2, \quad (27)$$

and cannot give a nontrivial exact value, since $f(\epsilon, 0) = 1$. The symmetry (20) in the new variables means that

$$f(\epsilon, z) = f(-\epsilon, -z), \quad f(-\epsilon, z) = f(\epsilon, -z). \quad (28)$$

It follows from (28) that the even (f_s) and odd (f_a) parts of $f(\epsilon, z)$ relatively to ϵ coincide with the even (f^s) and odd (f^a) parts of $f(\epsilon, z)$ relatively to z . The simplest way to satisfy these properties corresponds to the following functional form of $f(\epsilon, z)$

$$f(\epsilon, z) = f(\epsilon z, \epsilon^2, z^2) \quad (29)$$

One can conclude from (29) that in this case an expansion of $f(\epsilon, z)$ near the point $\epsilon = z = 0$ does not contain terms linear in ϵ and z separately. The odd part f_a can be represented in the form

$$f_a(\epsilon, z) = 2\epsilon z \Phi(\epsilon, z) \quad (30)$$

where Φ is an even function of ϵ and z (the coefficient 2 in front of ϵz is chosen for further convenience).

The DR (24) in general case is not enough for a complete determination of f , but it poses some constraint on the possible form of f . There are various ways of its choice. The most known example, using the partition of f into symmetric and antisymmetric parts $f = f_s + f_a$, connects f_s and f_a

$$f_s^2 - f_a^2 = 1 - z^2. \quad (31)$$

Since this form of the DR is associated with "a parity" partition of function f one can call it "a parity representation" of the DR. It means that f_a and f_s considered at fixed z as the functions of ϵ satisfy to hyperbolic relation with a constant depending on z . The relation (31) allows to express $f(\epsilon, z)$ through its even or odd parts

$$f(\epsilon, z) = f_a + \sqrt{f_a^2 + 1 - z^2} = f_s \pm \sqrt{f_s^2 - 1 + z^2}. \quad (32)$$

For this reason it is enough to know only one of these two parts. Usually one prefers to choose an antisymmetric part as more simple. Using the first approximation in z for f_a in the weakly inhomogeneous case, which follows from the mean conductivity

$$\langle \sigma \rangle = \sigma_+(1 + 2\epsilon z), \quad f_a(\epsilon, z) = 2\epsilon z. \quad (33)$$

and substituting (33) into (32), one obtains the effective conductivity

$$\sigma_e(\epsilon, z) = \sigma_+ \left[2\epsilon z + \sqrt{(2\epsilon z)^2 + 1 - z^2} \right], \quad (34)$$

which coincides with σ_e of self-dual square RRL model in the EMA [2]. We will refer the expression (34) as the EMA expression for effective conductivity of

self-dual systems. For self-reciprocal systems there are also relations analogous to (31,32) with a change $f_s, f_a \rightarrow f^s, f^a$, but in this case the relations (29,30) do not take place. For this reason the EMA cannot be a good approximation for SR systems.

For a checking the symmetry properties and a comparison of different expressions for σ_e it is useful to consider also σ_e of RRL models on other isotropic lattices, triangle and honeycomb. The EMA expression for σ_e of the binary lattice RRN models is [2]

$$\begin{aligned} \sigma_e &= \frac{1}{q-2} \left\{ \mathbb{I} + \sqrt{\mathbb{I}^2 + 2(q-2)\sigma_1\sigma_2} \right\}, \\ \mathbb{I} &= \left[\left(\frac{q}{2}x - 1 \right) \sigma_1 + \left(\frac{q}{2}(1-x) - 1 \right) \sigma_2 \right] \end{aligned} \quad (35)$$

where q is a number of bonds coming in (or outgoing from) one lattice site. In terms of z and ϵ it has the following form

$$\begin{aligned} \sigma_e &= \frac{\sigma_+}{q-2} \left\{ q \left(\frac{q-4}{2q} + \epsilon z \right) + \mathbb{I}^{1/2} \right\}, \\ \mathbb{I} &= [(q/2)^2 + q(q-4)\epsilon z + (q^2\epsilon^2 - 2(q-2))z^2] \end{aligned} \quad (36)$$

which has the necessary symmetry property (29). One can check that the corresponding $\sigma_e^{(tr)}$ ($q=6$) and $\sigma_e^{(hon)}$ ($q=3$) satisfy also the RR

$$\sigma_e^{tr}(\epsilon, z)\sigma_e^{hon}(-\epsilon, z) = \sigma_1\sigma_2.$$

As it was remarked earlier, some properties of RRN models differ from the properties of continuous media. For this reason, one can hope that the other possible representations for f compatible with the DR can produce other approximate expressions for σ_e , which will turn out more suitable for models with other inhomogeneity structures. Another reason to analyse the duality relation is connected with the known fact, that usually the systems with such symmetry have some additional, hidden, properties, permitting to obtain more information about function under consideration. Moreover, in some simple cases they can help to solve problem exactly (see, for example, [15]). Bearing this in mind, we will investigate various consequences of the duality relation in more details.

3. Duality, symmetries and perturbation expansions

We start with the series expansion of $f(\epsilon, z)$ on z . For every fixed z ($0 \leq z < 1$) a function f must be a monotonous function of ϵ . Since a homogeneous limit $z \rightarrow 0$ is a regular point of f it will be very useful to consider a series expansion of f in z

$$f(\epsilon, z) = \sum_0^\infty f_k(\epsilon)z^k/k!, \quad (37)$$

where due to boundary conditions (25')

$$f_0 = 1, \quad f_1(\epsilon) = 2\epsilon. \quad (38)$$

It is worth to note here that: 1) similar expansion was already considered up to third order (see, for example, [16]) and 2) the expansion (38) is connected with the weak disorder expansion for RRN models in moments of fluctuations $\delta\sigma$ [11]. We will show that our expansion is much simpler, since it treats the variables z and ϵ independently. From this point of view the expansion on moments of $\delta\sigma$ is an expansion on rather complicated functions of z and ϵ .

Substituting the expansion (37) into (26) one obtains the following results:

- (1) in the second order in z it reproduces the universal formula (12), thus the latter can be considered as a consequence of the duality relation;
- (2) in general case, there are the recurrent relations between f_{2k} and f_{2k-1} , corresponding to the connection (31) and expressing the even terms through the odd ones;
- (3) $f_{2k+1}(\epsilon)$ are odd polynomials of ϵ of degree $2k+1$ and $f_{2k}(\epsilon)$ are even polynomials of ϵ of degree $2k$ in agreement with (29).

Taking into account boundary conditions (25'), the exact value (26') and the symmetry property (29), one can show that the coefficients f_k must have the next form

$$\begin{aligned} f_{2k+1}(\epsilon) &= \epsilon(1 - 4\epsilon^2)g_{2k-2}(\epsilon), \quad k \geq 1, \\ f_{2k}(\epsilon) &= (1 - 4\epsilon^2)h_{2k-2}(\epsilon), \quad k \geq 1, \end{aligned} \quad (39)$$

where $g_{2k-2}(\epsilon)$ and $h_{2k-2}(\epsilon)$ are some even polynomials of the corresponding degree, and free terms of $h_{2k-2}(\epsilon)$ coincide with the coefficients in the expansion of the exact value (26')

$$\sqrt{1-z^2} = 1 - z^2/2 - z^4/8 - z^6/16 - z^8/128 - z^{10}/256 + \dots \quad (40)$$

The parity representation gives the relations, expressing f_{2k} with even indices through f_k with lower indices. For example,

$$f_4 = 4f_1f_3 - 3f_2^2 = (1 - 4\epsilon^2)[(8g_0 + 12)\epsilon^2 - 3], \quad (41)$$

$$f_6 = 6f_1f_5 - 15f_2f_4 + 10f_3^2. \quad (42)$$

It follows from (39) that f_3 is completely determined up to overall factor number g_0 . Since f_4 is determined through lower f_k ($k = 1, 2, 3$) it is also determined by the coefficient g_0 . In the EM approximation the series (21) is defined completely and takes very simple form, since in this case all $g_{2k-2} = 0$ ($k > 1$) and $f_{2k}(\epsilon) \sim (1 - 4\epsilon^2)^k$.

It is useful also to write out the series expansions for other 2D isotropic RRL models in the EM approximation. The expansion of (36) in z up to the 6-th order terms gives

$$\sigma_e = \sigma_+ \left\{ 1 + 2\epsilon z - \frac{2}{q}(1 - 4\epsilon^2)z^2 + \frac{4\epsilon(1 - 4\epsilon^2)(8 - 6q + q^2)}{q^2(q - 2)}z^3 \right.$$

$$\begin{aligned}
& - \frac{4(1-4\epsilon^2)(q-2+2\epsilon^2(20-10q+q^2))}{q^3} z^4 + \\
& + \frac{8\epsilon(1-4\epsilon^2)}{q^4(q-2)} (8-6q+q^2)(3(q-2)+2\epsilon^2(28-14q+q^2))z^5 - \\
& - \frac{16(1-4\epsilon^2)}{q^5} ((q-2)^2+2\epsilon^2(-112+112q-34q^2+3q^3) + \\
& + 2\epsilon^4(672-672q+224q^2-28q^3+q^4)) z^6 \}
\end{aligned} \tag{43}$$

It has the necessary structure following from the general properties. Note that the two first terms coincide with the universal (12) only for self-dual square RRL. One can see also from (43) that the higher odd terms (starting from the 3-nd order term) depend on the lattice structure and do not vanish except the self-dual case of the square lattice (where this takes place due to the factor $(8-6q+q^2)$).

In order to find higher order terms for square RRL model one may compare the expansion (37) with known results in series expansion in powers of conductivity fluctuations $\delta_k = \langle (\delta\sigma)^k \rangle$ [11]. Its coefficients are found up to 6-th order and it was shown that for 2D square RRL model, due to the duality relation, they begin to differ from the corresponding EMA coefficients only in the 5-th order

$$\begin{aligned}
\sigma_e^{sq} = & \langle \sigma \rangle \left(1 - \frac{\delta_2}{2} + \frac{\delta_3}{4} - \frac{\delta_4}{8} + \frac{\delta_5 + J_1 \delta_2 \delta_3}{16} \right. \\
& \left. - \frac{[\delta_6 + (3J_1 - 1)\delta_2 \delta_4 + (2J_1 - 1)\delta_3^2 - (3J_1 - 2)\delta_2^3]}{32} \right), \tag{44}
\end{aligned}$$

where $J_1 = \simeq 1.092958179$ is some lattice integral of the square lattice Green function. The EM approximation corresponds to $J_1 = 1$.

For a comparison of the general expansion (44) with (37) one needs the explicit expressions for the higher moments of $\delta\sigma$. After some algebra one can show that the moments of binary systems with the distribution function (7) have in terms of variables σ_- and x (or z and ϵ) the following form

$$\begin{aligned}
\langle (\Delta\sigma)^k \rangle &= x(1-x)P_{k-2}(x)(\sigma_-)^k = (\sigma_+)^k(1-4\epsilon^2)/4P_{k-2}(\epsilon)z^k, \\
P_{k-2}(x) &= (1-x)^{k-1} + (-1)^k x^{k-1}, \\
P_{k-2}(\epsilon) &= (1/2-\epsilon)^{k-1} + (-1)^k (\epsilon+1/2)^{k-1}
\end{aligned} \tag{45}$$

One can see from (45) that z -dependence of the moments is factorised. Moreover, it follows from the explicit expression for the polynomial P_{k-2} that for odd $k = 2m+1$, due to the symmetry of this polynomial relative interchange $x \leftrightarrow 1-x$, it has additional general factor $(1-2x) = -2\epsilon$ in accordance with (39). Thus, we see that the series expansion for square RRL model in δ_k from [11] has in terms of variables z and ϵ the necessary structure, following from symmetry and duality. The obvious advantage of series expansion (38) is that it

treats the variables z and ϵ separately. The explicit forms of the polynomials P_{k-2} at $k \leq 6$ are

$$\begin{aligned}\langle \Delta^3 \rangle &= 4\epsilon(1-4\epsilon^2)\sigma_-^3, & \langle \Delta^4 \rangle &= 4(1-4\epsilon^2)(1-3x+3x^2)\sigma_-^4, \\ \langle \Delta^5 \rangle &= 4\epsilon(1-4\epsilon^2)(1-2x+2x^2)\sigma_-^5, \\ \langle \Delta^6 \rangle &= 2^4(1-4\epsilon^2)(1-5x+10x^2-10x^3+5x^4)\sigma_-^6.\end{aligned}\quad (46)$$

Using the equality (33) for $\langle \sigma \rangle$ and expanding the terms of the series (44) in z , one can obtain the series (37) for the square RRL model up to 6-th order. In particular, in the EMA one obtains the corresponding series (43). Since the series for square RRL model begin to differ from the corresponding EMA in 5-th order it is enough to find only 5-th and 6-th order terms. They are

$$g_2 = -30j(1-4\epsilon^2), \quad h_4 = -45(1-4\epsilon^2)[1-4(1-2j)\epsilon^2], \quad (47)$$

where $j = J_1 - 1 = 0.092958179 \ll 1$. One can check that these formulas satisfy relations (41,42).

Thus, we have obtained the series in z for σ_e of self-dual square RRL model up to 6-th order. Since it coincide with the EMA up to 4-th order, one can say that they belong to the same class of functions with $g_0 = 0$. As follows from (43) the effective conductivity for isotropic RRL models in the EM approximation depends on the lattice structure and can have $g_0 \neq 0$. Then the next question appears: is the effective conductivity of the continuous self-dual systems universal and always belongs to the EMA class or there are SD systems with other functional forms of σ_e ? The answer on this question will be obtained in the subsequent sections.

4. Exponential representation and new approximate expressions

Thus we have seen that the arbitrariness of f of self-dual systems is strongly reduced by boundary conditions and the exact value (26'), and that the third and fourth order terms are determined only up to one constant g_0 , which equals 0 for the square RRL model and in the EM approximation. The question about the universality reduces in this particular case to the next one: is the equality $g_0 = 0$ true for all self-dual random inhomogeneous systems or it depends on the structure of random inhomogeneities? If the value of g_0 depends on random structure, then the second question appears: can any other additional information about the function f of these systems determine the constant g_0 or even the whole function, as it was in case of the EMA? Since the EMA is connected with the parity representation (31) and a mean conductivity approximation (33) for odd part, one needs to know what kind of other representations for f can satisfy the duality relation (26). In order to answer on this question it is convenient in the case $z \neq 1$ to pass from f to $\tilde{f} = f/\sqrt{1-z^2}$. Then

$$\tilde{f}(\epsilon, z)\tilde{f}(-\epsilon, z) = 1 = \tilde{f}(\epsilon, z)\tilde{f}(\epsilon, -z). \quad (48)$$

The DR (48) assumes other possible functional forms of $\tilde{f}(\epsilon, z)$. Taking into account (29), one can write out the exponential representation for \tilde{f}

$$\tilde{f}(\epsilon, z) = \exp(\epsilon z \phi(\epsilon, z)), \quad (49)$$

where $\phi(\epsilon, z)$ is some even function of its arguments. It is easy to see that it automatically satisfies eq.(48). Now let us find out is it possible to define g_0 from some additional information about function ϕ . For this we consider two simple ansatzes for $\phi(\epsilon, z)$, analogously to the parity representation (31) and the EM approximation.

1) In the first case we will assume that $\phi(\epsilon, z)$ depends only on z . This corresponds to the exponential dependence on concentration. Such dependence takes place often in disordered systems [17]. Expanding the corresponding functions f in series one can check after some algebra that now all polynomial coefficients can be determined unambiguously! The series for f_a up to 5-th order has the form

$$f_a = \sum_0^2 f_{2k+1} \frac{z^{2k+1}}{(2k+1)!}, \quad f_{2k+1} = \epsilon(1 - 4\epsilon^2)g_{2k-2}, \quad k = 1, 2, \\ g_0 = -1, \quad g_2 = -(11 + 4\epsilon^2). \quad (50)$$

Another way to see this is to apply the boundary conditions (25') directly to the function (49). Then one obtains the whole function

$$\phi(z) = 1/z \ln \frac{1+z}{1-z}, \quad \tilde{f}(\epsilon, z) = \left(\frac{1+z}{1-z} \right)^\epsilon. \quad (51)$$

The series expansion of (51) reproduces exactly the corresponding expansion from (50). Note that now $g_0 \neq 0$. The even part f_s can be found from (41,42).

2) In the second case we will assume that $\phi(\epsilon, z)$ depends only on the combination ϵz . This corresponds to the dependence of σ_e only on mean conductivity and/or mean resistivity, since $\langle \sigma^{\pm 1} \rangle \sim (1 \pm 2\epsilon z)$. Expanding the corresponding functions \tilde{f} in series one can see that again all polynomial coefficients can be determined completely. The coefficients for f_a up to 5-th order are

$$g_0 = -3, \quad g_2 = -15(1 + 12\epsilon^2) \quad (52)$$

In this case one can also apply the boundary conditions (25') directly to the function (49). Then one obtains

$$\phi(\epsilon, z) = \frac{1}{2\epsilon z} \ln \frac{1+2\epsilon z}{1-2\epsilon z}, \quad \tilde{f}(\epsilon, z) = \left(\frac{1+2\epsilon z}{1-2\epsilon z} \right)^{1/2}. \quad (53)$$

The series expansion of (53) also coincides exactly with the corresponding expansion represented in (52).

It is worth to note that an analogous exponential representation can be obtained for self-reciprocal, but nonsymmetric systems. In this case only the

second equality in (48) is fulfilled and the exponential representation has the form

$$\tilde{f}(\epsilon, z) = \exp(z\phi(\epsilon, z)), \quad (54)$$

where $\phi(\epsilon, z)$ is an odd function of z . The boundary conditions (25') cannot now determine it completely. They only give

$$\phi\left(\pm\frac{1}{2}, z\right) = \pm\frac{1}{2z} \ln\left(\frac{1+z}{1-z}\right).$$

The simplest way to satisfy these boundary conditions is to assume that the interpolating function at arbitrary ϵ has factorized dependencies on ϵ and z (it is an analog of the assumption 1) in the SD case)

$$\phi(\epsilon, z) = \frac{\alpha(\epsilon)}{z} \ln\left(\frac{1+z}{1-z}\right), \quad (55)$$

here $\alpha(\epsilon)$ is an arbitrary function except its boundary values

$$\alpha(\pm 1/2) = \pm 1/2.$$

For self-dual systems $\alpha(\epsilon) = \epsilon$, and just this equality ensures the exact Keller-Dykhne value (22,26') at $\epsilon = 0$. Formally, the SR relation admits the self-dual solution (52) in nonsymmetric case too, but, as we will show below in section 7, (52) works well only for ϵ , belonging to small regions near the boundary values $\epsilon = \pm 1/2$ (or at small concentrations of one of the phases $x \ll 1, 1-x \ll 1$). Its applicability at intermediate values of ϵ depends on how close is a non self-dual system to the self-dual one.

Thus we have shown that under some simple (but physically acceptable) additional assumptions about a function $f(\epsilon, z)$ one can obtain the new approximate (a mean field like) expressions for f . Their physical meaning and properties will be discussed in two subsequent sections where we will present two simple models having the effective conductivity just of two forms found above. Here we only want to note that it is very important for further progress in this direction to find any other explicit solutions. In general case, assuming an expansion of ϕ in double series in z and ϵ ,

$$\phi(\epsilon, z) = \sum_{k,l=0}^{\infty} \phi_{kl} \frac{z^{2k}}{k!} \frac{\epsilon^{2l}}{l!}$$

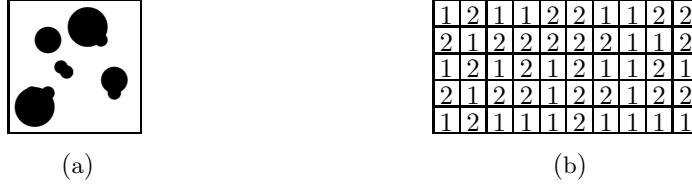
one can show that now again the boundary conditions (25') cannot define all coefficients completely. For example, f_3 and f_4 contain one free parameter ϕ_{10} : $g_0 = 6(\phi_{10} - 1)$.

5. Finite maximal scale averaging approximation

In this section we show that the solution (52) can describe σ_e of inhomogeneous systems with compact inclusions of one of the phases. Firstly, we

introduce a new composite method of a construction of random inhomogeneous systems. In framework of this approach one divides an averaging procedure on two steps, corresponding to the averaging on two different scales. Then, using this approximate averaging, which we will call as a finite maximal scale averaging approximation (FMSA), we will obtain an additional equation for effective conductivity of inhomogeneous systems with compact inclusions. This approximation can be considered as some modification of the mean field approximation and it is applicable to the random inhomogeneous systems with a finite maximal scale of the inhomogeneities. Below in this section we will use temporally, for clarity, the concentration variable x instead of ϵ . Let us consider planar two-phase randomly inhomogeneous system. It is easy to see that, in general, its effective conductivity will depend on a scale l of a region over which an averaging is done. This takes place due to the possible existence of different characteristic scales in the inhomogeneous medium. In the most general case there will be a whole spectrum of these characteristic scales. This spectrum can change from discrete finite one till continuous infinite one and will define the inhomogeneity structure of the system. For this reason this spectrum can depend on concentration x . It is obvious that the effective conductivity of the system can depend on this spectrum as a whole as well as on which area an averaging is fulfilled over. Suppose, for the simplicity, that the randomly inhomogeneous structure of our system has a scale spectrum with the maximal scale $l_m(x)$, which is finite for all x in the region $1 \geq x > 1/2$ (or $1 - x$ in the region $0 \leq 1 - x < 1/2$). Let us assume that we know an exact formula for $\sigma_e(x, z)$ of this system, which is applicable from scales $l > l_m$. It means that this formula for $\sigma_e(x, z)$ takes place after the averaging over regions with a mean size $l \gtrsim l_m$ and does not change for all larger scales $l \gg l_m$. Now we deduce an approximate functional equation for this $\sigma_e(x, z)$. To do this let us consider the auxiliary square lattice with the squares of length $l_L \gg l_m$ and suppose that they have the effective conductivities corresponding to different values of the concentrations x_1 and x_2 with equal probabilities $p = 1/2$ (see Fig.2).

After the averaging over the scales $l \gtrsim l_L$ one must obtain on much larger scales $l \gg l_L$ the same effective conductivity, but corresponding to another concentration $x = (x_1 + x_2)/2$. This is possible due to the similar random structure of different squares and due to the conjectures that in this model: (1) there are only two maximal characteristic scales $l_m(x_i) \equiv l_i$ ($i = 1, 2$) and the lattice square size $l_L \gg l_i, l_m(x)$, (2) the averaging procedures over these scales do not correlate (or weakly correlate) between themselves. Thus, for a compatibility, all concentrations must be out of small region around critical concentration x_c , where l_i or $l_m(x)$ can be very large. We will call further this set of the assumptions the finite maximal scale averaging approximation (FMSA approximation). It can be implemented for systems with compact inhomogeneous inclusions with finite l_m . From the other side the effective conductivity of the auxiliary plane on scales $l \gg l_L$ must be determined by the universal Keller – Dykhne formula (22). Thus we obtain the next functional equation for the effective conductivity,



(a)

(b)

Figure 2: (a) An elementary square of the auxiliary lattice with compact inclusions, (b) the auxiliary lattice, the numbers 1,2 denotes squares with the corresponding concentrations x_1 and x_2 .

connecting $\sigma_e(x, z)$ at different concentrations,

$$\sigma_e(x, z) = \sqrt{\sigma_e(x_1, z)\sigma_e(x_2, z)}, \quad x = (x_1 + x_2)/2, \quad (x_i, x \neq x_c). \quad (56)$$

It must be supplemented by the boundary conditions (25'). This equation can be considered as a generalization of the duality relation (21), the latter being a particular case of (56) at $x_1 + x_2 = 1$. It follows from (56) that, due to the exactness of the duality relation, it really works for all concentrations x , except, maybe, a small region near $x = x_c$ and $z = 1$ (this region corresponds to the singular region of the percolation problem, see also below a discussion of the percolation limit). One can easily check that σ_e in low concentration limit, being a linear function of x , satisfies (56). Moreover, one can find an exact solution of this equation. It has an exponential form with a linear function of x (or ϵ)

$$\sigma_e(x, z) = \sigma_1 \exp(ax + b), \quad (57)$$

where the constants a, b can be determined from the boundary conditions

$$a = -b, \quad \exp b = \sigma_2/\sigma_1. \quad (57')$$

Substituting these coefficients into (57) one obtains

$$\sigma_e(x, z) = \sigma_1 (\sigma_2/\sigma_1)^{(1-x)} = \sigma_+ \sqrt{1-z^2} \left(\frac{1+z}{1-z} \right)^\epsilon. \quad (58)$$

The solution (58) satisfies symmetry relation (20) and can be represented in the exponential form

$$\sigma_e(x, z) = \sigma_+ \sqrt{1-z^2} \exp \left(\epsilon \ln \frac{1+z}{1-z} \right), \quad (58')$$

which coincides exactly with the case 1) from the section 4 with $\phi(\epsilon, z) = \frac{1}{z} \ln \frac{1+z}{1-z}$. Its odd part has a form

$$\tilde{f}_a(\epsilon, z) = \sinh(\epsilon z \phi(\epsilon, z)), \quad (59)$$

and automatically satisfies (31).

It is interesting to note that the form of the solution (58) means that in the considered approximation one has effectively an averaging of the $\log \sigma$ since it can be represented as

$$\log \sigma_e = \langle \log \sigma \rangle = x \log \sigma_1 + (1 - x) \log \sigma_2. \quad (60)$$

This coincides with a note made in [13] for the case of equal concentrations $x = 1/2$ and with analogous results obtained later for disordered systems in the theory of weak localization [18]. The solution (58) satisfies also the inequalities (11) due to the Hölder inequalities

$$\langle \sigma^{-1} \rangle^{-1} \leq \sigma_e(x, z) = \sigma_1^x \sigma_2^{(1-x)} \leq \langle \sigma \rangle. \quad (61)$$

One can check that (58) reproduces in the weakly inhomogeneous limit the universal Landau – Lifshitz expression (12). In the low concentration limit of the second phase it gives

$$\sigma_e(x, z) = \sigma_1 \left(1 + (1 - x) \log \frac{1 - z}{1 + z} + \dots \right), \quad 1 - x \ll 1, \quad (62)$$

what coincides with the Maxwell-Garnett formula in the weakly inhomogeneous case [1]. Note that the expansion (62) contains the coefficient logarithmically diverging in the limit $|z| \rightarrow 1$ ($\sigma_2 \rightarrow 0$). Such behaviour of the coefficients denotes the existence of a singularity in this limit as it must be due to the percolation singularity (see below a discussion of this percolation limit).

As follows from the arguments of the method used above, the FMSA can be applied also for approximate description not only of random systems, but also for any (regular as well as irregular) inhomogeneous models with compact inclusions of one phase into another. However, the explicit approximate expression (58) obtained for σ_e will work well in a wide range of parameters only for systems close to the self-dual ones (see the notes in the sections 4 and 7). For example, this expression describes σ_e of the Rayleigh model with strictly round inclusions not very well for intermediate concentrations (it gives smaller values) as it shows the comparison from section 7 (it can be expected also from Fig.1). But it will work much better when the shape of these inclusions is deformed so that the corresponding pictures of this model (Fig.1a and Fig.1d, Fig.1b and Fig.1c) become similar and symmetry property (20) is approximately fulfilled.

6. Random "parquet" model

Now we will show that the existence of different functional forms for the effective conductivity denotes a nonuniversal character of this value for self-dual two-phase systems with different random structures. We will construct another model of two-dimensional isotropic randomly inhomogeneous two-phase system, using the composition method introduced above, and find a mean field like expression for its effective conductivity $\sigma_e(x, z)$ in case of arbitrary phase concentrations x . The derived formula satisfies again to all necessary symmetries,

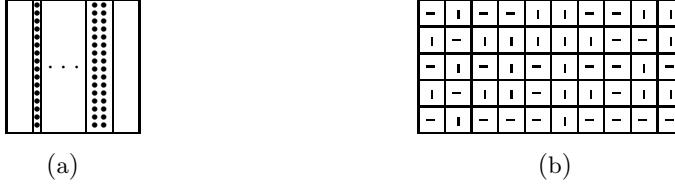


Figure 3: (a) An elementary square of the model with a vertical orientation, the dotted regions denote layers of the second phase; (b) a lattice of the model, the small lines on the squares denote their orientations.

including a self-dual one, and coincides with the example case 2) from section 4, realizing the second variant, when $\tilde{f}(x, z)$ depends (in this approximation) only on one combination of variables ϵz . This dependence is described by the function analytical at small values of this variable.

Let us consider the following planar model. There is a simple square lattice with the squares consisting of a random layered (or striped) mixture of two isotropic conducting phases with constant conductivities $\sigma_i, i = 1, 2$ and the corresponding concentrations x and $1 - x$. A schematic picture of such square is given in Fig.3.

The layered structure of the squares means that the squares have some preferred direction, for example along the layers. Let us suppose that the directions of different squares are randomly oriented (parallelly or perpendicularly) relative to the external electric field, which is directed along x axis. In order for system to be isotropic the probabilities of the parallel and perpendicular orientations of squares must be equal or (what is the same) the concentrations of the squares with different orientations must be equal $p_{||} = p_{\perp} = 1/2$.

Such lattice can model a random system consisting from mixed phase regions, which can be roughly represented on the small macroscopic scales as randomly distributed plots with the effective "parallel" and "serial" connections of the layered two-phase mixture (Fig.3). The lines on the squares denote their orientations.

This layered structure can appear, for example, on the intermediate scales when a random medium is formed as a result of the stirring of the two-phase mixture or as a result of appearance of randomly distributed regions of "striped phases" in oxide materials [3]. The corresponding averaged parallel and perpendicular conductivities of squares $\sigma_{||}(x)$ and $\sigma_{\perp}(x)$ are defined by well known formulas

$$\begin{aligned}\sigma_{||}(x) &= \langle \sigma \rangle = x\sigma_1 + (1-x)\sigma_2 = \sigma_+(1+2\epsilon z), \\ \sigma_{\perp}(x) &= \langle \sigma^{-1} \rangle^{-1} = \left(\frac{x}{\sigma_1} + \frac{1-x}{\sigma_2} \right)^{-1} = \sigma_+ \frac{1-z^2}{1-2\epsilon z}.\end{aligned}\quad (63)$$

Thus we have constructed the hierarchical random medium (in this case a two-level one). On the first level it consists from some regions (squares of two different types) of the random mixture of the two layered conducting phases with different conductivities σ_1 and σ_2 and arbitrary concentration. On the second level this medium is represented as a random parquet constructed from squares of two such types with different conductivities $\sigma_{||}$ and σ_{\perp} , depending nontrivially on concentration of the initial conducting phases, and randomly distributed with the same probabilities $p_i = 1/2$ (Fig.3). This representation allows us to divide the averaging process approximately into two steps (firstly averaging over each square and then averaging over the lattice of squares) and implement on the second step the exact formula (22). This can be considered as some modification of the FMSA approximation. As a result one obtains for the effective conductivity of the introduced random "parquet" model the following formula, which is applicable for arbitrary concentration

$$\sigma_e(\epsilon, z) = \sqrt{\langle \sigma \rangle \langle \sigma^{-1} \rangle^{-1}} = \sigma_+ \sqrt{1 - z^2} \tilde{f}(\epsilon, z), \quad \tilde{f}(\epsilon, z) = \left[\frac{1 + 2\epsilon z}{1 - 2\epsilon z} \right]^{1/2}, \quad (64)$$

This function has all necessary properties and satisfies SD equation (26), inequalities (11) and the symmetry (20). It coincides with the solution 2) from section 4 and has another possible functional form, automatically satisfying the duality relation (48)

$$\tilde{f}(\epsilon, z) = B(\epsilon, z)/B(-\epsilon, z) \quad (65)$$

with a function $B(\epsilon, z) = [1 + 2\epsilon z]^{\frac{1}{2}}$, which depends only on the combination ϵz . Strictly speaking, this form is not independent, since (65) can be also represented in the exponential form

$$\tilde{f}(\epsilon, z) = \exp(\phi(\epsilon, z))$$

where $\phi(\epsilon, z)$ is an odd function of ϵ

$$\phi(\epsilon, z) = \ln B(\epsilon, z)/B(-\epsilon, z).$$

It is interesting to compare this formula with the known asymptotic formulas. Let us consider firstly its behaviour for small phase concentrations.

(a) In case of small concentration of the first phase $x \ll 1$ one gets

$$\sigma_e(x, z) \simeq \sigma_2 \left(1 + \frac{2xz}{1 - z^2} \right). \quad (66)$$

It follows from (66) that an addition of small part of the first higher conducting phase increases an effective conductivity of the system as it should be.

(b) In the opposite case of small concentration of the second phase $1 - x \ll 1$ one obtains

$$\sigma_e(x, z) \simeq \sigma_1 \left(1 - \frac{2(1-x)z}{1 - z^2} \right), \quad (67)$$

i.e. an addition of the phase with smaller conductivity decreases σ_{eff} . It is worth to note that both these expressions for arbitrary values of the conductivities σ_1 and σ_2 differ from the Maxwell-Garnett formula and coincide with

it only in the weakly inhomogeneous case $z \ll 1$. It must be not surprising because a form of the inclusions of the second phase in this model has completely different, layered, structure. In the low concentration expansion as well as directly in formula (64) one can see again that the divergencies appear in the limit $z \rightarrow 1$.

(c) In case of almost equal phase concentrations $x = 1/2 + \epsilon$, $\epsilon \ll 1$ one obtains

$$\tilde{f}(\epsilon, z) \simeq 1 + 2\epsilon z, \quad \sigma_{eff}(\epsilon, z) \simeq \sigma_+ \sqrt{1 - z^2} (1 + 2\epsilon z). \quad (68)$$

The Keller – Dykhne formula (22) is reproduced for equal concentrations.

The antisymmetric part of \tilde{f} has the following form

$$\tilde{f}_a(\epsilon, z) = \frac{\tilde{f}^2(\epsilon, z) - 1}{2\tilde{f}(\epsilon, z)} = \frac{2\epsilon z}{[1 - 4\epsilon^2 z^2]^{1/2}}. \quad (69)$$

It follows from formula (41) that for $\epsilon \ll 1$ and (or) $z \ll 1$ the odd part f_a coincides in the first order with the corresponding expression from the effective medium theory. The corresponding function Φ is

$$\Phi(\epsilon, z) = \frac{\sqrt{1 - z^2}}{[1 - 4\epsilon^2 z^2]^{1/2}}. \quad (70)$$

In other words it is an analytic function of ϵz near $\epsilon z = 0$. Basing on this formula one can conjecture that an exact expression for $\tilde{f}_a(\epsilon, z)$ of this model will have a similar structure in general case with $\sigma_i \neq 0$ or $1 - z \neq 1$. One must note that at the same time the formula (64) does not satisfy the equation (56) except of the trivial case $x_1 = x_2$.

7. Comparison of different approximations

For the comparison of the different expressions of the effective conductivity we have constructed 3D plots of $f(\epsilon, z)$ in the EM approximation, in the FMSA approximation and for the random "parquet" model in FMSA-like approximation (fig.4,5a). For a clarity they are represented at $z \leq 0$ ($\sigma_1 \leq \sigma_2$).

It follows from fig.4 and fig.5a that all three formulas for σ_e , despite of their various functional forms, differ from each other weakly for $z \lesssim 0.8$ due to very restrictive boundary conditions (25') and the exact Keller-Dykhne value. This range of z corresponds approximately to the ratio $\sigma_2/\sigma_1 \sim 10^{-1}$. For the smaller ratios a difference between these functions become distinguishable.

It is useful also to compare σ_e in the FMSA approximation with σ_e^{rm} of the Rayleigh model (RM), constructed numerically [14]. Fig.5b shows f in the FMSA approximation as a function of x and $h = \frac{1-z}{1+z}$ for the direct comparison with the numerical results for RM from [14]. σ_e^{rm} satisfies the self-reciprocity relation (17), but does not satisfy the duality relation (24). For this reason σ_e^{rm} at adjoint concentrations are not related. The critical concentration x_c of the RM in the percolation limit $z \rightarrow 1$ is $x_c = 1 - \pi/4 \approx 0.215$ [14]. It differs strongly from the critical concentration of self-dual systems $x_c = 1/2$. By

compairing fig.3 from [14] and our figs.4b,5b one can see that for $z \lesssim 0.8$ both σ_e are very similar and close to each other. For $z \geq 0.8$ they begin to differ, our σ_e becomes smaller than σ_e^{rm} for concentrations in the range $0.2 \leq x \leq 0.8$ due to a fact, that σ_e from (58) must follow the exact value (22) at $x = 1/2$. At the same time both conductivities remain close for concentrations in small regions near $x = 1$ and $x = 0$. This behaviuor can be explained by the difference in the geometry and symmetry of both systems, which becomes significant in this range of concentrations. For this reason one can expect, that for the RM with round inclusions deformed in a such way that the approximate symmetry (20) appears, its σ_e^{rm} will become more similar to σ_e from (58).

8. Percolation limit

Now let us consider in the more details the derived formulas for $\sigma_e(\epsilon, z)$ in case when $\sigma_2 \rightarrow 0(z \rightarrow 1)$. It is clear that for regularly inhomogeneous medium one can always construct such distribution of the conducting phase that $\sigma_e(\epsilon, 1)$ will differ from zero for all $1/2 \geq \epsilon > -1/2$. But in the case of randomly inhomogeneous medium the limit $\sigma_2 \rightarrow 0$ is equivalent to the well known percolation problem [17]. In terms of z it corresponds to the limit $z \rightarrow 1$ and is also similar to the ideal conductivity limit $\sigma_1 \rightarrow \infty$. Strictly speaking, an implementation of the duality transformation (14) is not obvious in this case. However, if one supposes that the dual symmetry relation (24) fulfills in this limit too due to a continuity then it follows from (24) that

$$\sigma_e(\epsilon)\sigma_e(-\epsilon) = 0. \quad (71)$$

The relation (71) does not contradict to the known basic results of the percolation theory that $\sigma_e(\epsilon) = 0$ for $\epsilon \leq 0$ and $\sigma_e(\epsilon) \neq 0$ for $\epsilon > 0$. Moreover it follows from the general formula (31) that in this case $\sigma_s = |\sigma_a|$ and $\sigma_e(\epsilon) = \sigma_a + |\sigma_a|$. It gives

$$\sigma_e(\epsilon) = \begin{cases} 0, & \epsilon \leq 0, \\ 2\sigma_a, & \epsilon > 0. \end{cases} \quad (72)$$

This means that a behaviour of $\sigma_e(\epsilon)$ in the percolation theory is completely determined by its odd part. From a general discussion of the functional structure of the conductivity of the two-dimensional randomly inhomogeneous systems and experimental and numerical results it is known that in the percolation limit the effective conductivity σ_e must have a nonanalytical behaviour near the percolation edge $x_c = 1/2$ or at small $\epsilon > 0$ [17]

$$\sigma_e(\epsilon) \sim \sigma_1(x - x_c)^t \sim \sigma_1\epsilon^t, \quad (73)$$

where a critical exponent of the conductivity t is slightly above 1 and can be represented in the form $t = 1 + \delta$. Since the values of this exponent found by the numerical calculations are confined to be in the interval $(1.10 - 1.4)$ [17],

then δ have to be small and belongs to the interval $(0.1 - 0.4)$. It follows from general formula (72) that one must have

$$f_a(\epsilon, z) \xrightarrow[z \rightarrow 1, \epsilon \rightarrow 0_+]{} \epsilon^t. \quad (74)$$

It means that the function $\Phi(\epsilon, z)$ from (30) has at small ϵ some crossover on z under $z \rightarrow 1$ from a regular (analytical) behaviour to a singular, powerlike one. At the moment an exact form of this crossover is unknown. For example, it can be of the form

$$\Phi(\epsilon, z) \sim (\Phi_0(z^2) + \Phi_1(z^2)\epsilon^2)^{\delta(z)/2}, \quad z \rightarrow 1, \quad (75)$$

where $\Phi_0(z^2) \rightarrow 0$, $\Phi_1(z^2) \rightarrow \Phi_1 \neq 0$ and $\delta(z) \rightarrow \delta \neq 0$ when $z \rightarrow 1$. Using (75) together with (30) and (32) one can show that this crossover takes place at $1 - z \sim \epsilon^{2t}$ in agreement with result obtained from the similarity hypotheses [19, 20].

It follows from the formulas for σ_e obtained in the sections 4-6, that one gets always $\sigma_e \rightarrow 0$ in the limit $\sigma_2 \rightarrow 0$, except the region near $x = 1$. It means that these formulas obtained in the FMSA approximation are not valid in the limit $\sigma_2 \rightarrow 0$. This is confirmed by the appearance of the divergencies in the expansions of σ_e in small concentrations in the limit $z \rightarrow 1$. This fact is a consequence of the assumptions made in the used approximations. For example, in case of the model of the layered (or striped) squares, where a deviation from the percolation type behaviour in the limit $z \rightarrow 1$ is more obvious, this is due to the "closing" (or "locking") effect of the layered structure in the adopted approximation in the limit $\sigma_2 \rightarrow 0$. In order to obtain a finite conductivity in this model above threshold concentration x_c one needs to take into account the correlations between adjacent squares. It is easy to show that near the threshold an effective conductivity is determined by random conducting clusters formed out of the crossing random layers from neighboring elementary squares. As is well known, the mean size of these clusters diverges near the percolation threshold [17] and for this reason the FMSA type approximation cannot be applicable for the description of σ_e in the limit $z \rightarrow 1 (\epsilon > 0)$ and of the percolation problem. The divergencies of the expressions in the percolation limit mean strong changes of these expressions, which must result in a formation of the (widely believed) universal critical behaviour (73). It follows from figs.4,5 that the EM approximation overestimates σ_e , and both other formulas underestimate it in the region $z \rightarrow 1, \epsilon > 0$. We hope to investigate this limit in detail in other papers.

9. Conclusions

Thus we have studied the possible functional forms of the effective conductivity of the two-phase SD system at arbitrary values of concentrations. A new approximate functional equation for σ_e , applicable for inhomogeneous self-dual systems and generalizing the duality relation, was deduced in the FMSA

approximation and its solution was found. We have constructed also a hierarchical ("parquet") model of the random inhomogeneous stripe type medium and have found its effective conductivity in the similar approximation at arbitrary phase concentrations. All expressions for the effective conductivity have the different functional forms. They

- (1) satisfy the self-dual symmetry and all necessary inequalities,
- (2) reproduce the general formulas for σ_e in the weakly inhomogeneous case.

All these results confirm a conjecture that, in general, σ_e already of two-phase self-dual systems may be a nonuniversal function and can depend on some details of the structure of the randomly inhomogeneous regions [21]. Analogous conclusions for SD systems with $N = 3$ phases were done earlier in the paper [22], where a possibility to find a generalization of the exact Keller-Dykne formula for case $N = 3$ was investigated numerically.

The obtained explicit formulas can be used for an approximate description of the effective conductivity of some real (as regular as random) inhomogeneous systems like a corresponding formula of the EM approximation. Another important property of these formulas is connected with a fact that they admit a generalization on a case of heterophase systems with arbitrary number of phases N (see [23] and my forthcoming paper). At the same time the formulas (58),(64) conserve their simple form for arbitrary N , while the EMA expression for σ_e becomes very complicated function even for $N = 3$.

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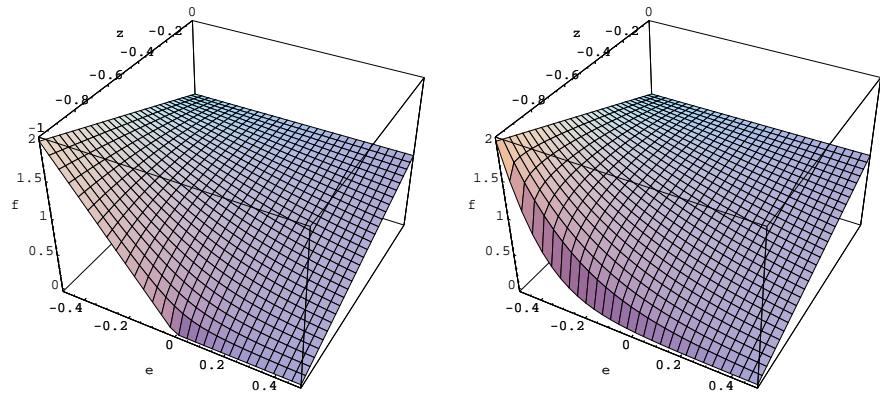


Figure 4: Plots of $f(\epsilon, z)$ in : a) the EM approximation, b) the FMSA approximation.

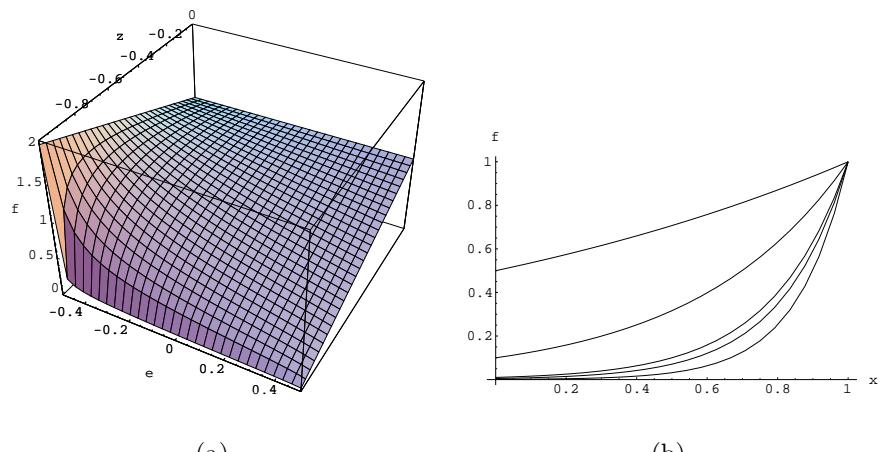


Figure 5: a) Plot of $f(\epsilon, z)$ of random "parquet" model in FMSA-like approximation,
b) Plot of $f(x, h)$ in FMSA approximation at $h = 0.5, 0.1, 0.01, 0.005, 10^{-3}$.